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## Technical Memorandum 79542

# Grain Formation in Astronomical Systems: A Critical Review of Condensation Processes

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**Bertram Donn**

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National Aeronautics and  
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**Goddard Space Flight Center**  
Greenbelt, Maryland 20771



# GRAIN FORMATION IN ASTRONOMICAL SYSTEMS: A CRITICAL REVIEW OF CONDENSATION PROCESSES

BY

Bertram Donn  
Laboratory for Extraterrestrial Physics  
NASA/Goddard Space Flight Center  
Greenbelt, Maryland 20771

## ABSTRACT

An analysis is presented of the assumptions and the applicability of the three theoretical methods for calculating condensations in cosmic clouds where no pre-existing nuclei exist. The three procedures are: thermodynamic equilibrium calculations, nucleation theory and a kinetic treatment which would take into account the characteristics of each individual collision. Thermodynamics provides detailed results on the composition temperature and composition of the condensate provided the system attains equilibrium. Because of the cosmic abundance mixture of elements, large supersaturations in some cases and low pressures, equilibrium is not expected in astronomical clouds. Nucleation theory, a combination of thermodynamics and kinetics, has the limitations of each scheme. Kinetics, not requiring equilibrium, avoids nearly all the thermodynamics difficulties but requires detailed knowledge of many reactions which thermodynamics avoids. It appears to be the only valid way to treat grain formation in space. To do this will require experiments on condensation that will provide an understanding of the process as it occurs in astronomical systems and also will provide the data on cross-sections, rates and related characteristics of the separate steps. A review of experimental studies concludes the paper.

# GRAIN FORMATION IN ASTRONOMICAL SYSTEMS: A CRITICAL REVIEW OF CONDENSATION PROCESSES

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## INTRODUCTION

Analyses of the cloud collapse phase of star formation, accumulation of solid grains into larger solar system objects, trapping of elements later incorporated into larger objects, thermal balance and temperature of clouds, are among the processes in which cosmic grains play an important role. The formation mechanism of grains largely determine their composition, concentration as a function of time and position, radiative characteristics and accumulation probability. Consequently, a review of the status of formation mechanism of grains is very pertinent to this conference.

In this paper I treat the case of condensation from a gas in the absence of any pre-existing grains or nuclei upon which condensation can occur. Some of the analysis is expected to apply to condensation on grains already present. The paper is not a summary of the 'state-of-the-art' in describing the formation of grains. It is, rather, an attempt to examine the basic assumptions of the various procedures used to describe condensation and to examine their applicability to astronomical systems.

The case of heterogeneous nucleation on ions or of growth on pre-existing grains is not discussed here. Ions may be important for stellar or circumstellar condensation. Conclusions about the nature of the grain and the mechanism are not expected to be appreciably changed. Nucleation would occur at higher temperatures or lower concentrations than with a neutral vapor. This problem will be taken up at a later time. Very small grains may be present in some of the dense clouds considered here which formed from low density clouds.

If interstellar grains survive the collapse of a diffuse cloud, grains less than  $100 \text{ \AA}$  are probably present (Greenberg, 1973). Some aspects of the growth of grains are given in Greenberg's report at the conference. The process, in outline, is described by Greenberg (1973) and by Field (1975) but detailed effects of gas-grain interaction (Hirth and Pound, 1963) are not included. As part of the Goddard study of cosmic grains, an analysis of this problem is intended.

Three mechanisms have been used to describe condensation. In historical order they are: (1) thermodynamics, Wildt (1933), (2) kinetics, Lindblad (1935), and (3) nucleation theory (Kamijo, 1966, 1968; Donn et al., 1968) which is a combination of thermodynamics and kinetics. These analyses have subsequently undergone considerable development. A comprehensive review of the thermodynamic procedure has been given by Grossman and Larimer (1974). A brief review of thermochemistry and nucleation theory has recently been published by Salpeter (1977).

Kinetic analyses require the most detailed input data and few general treatments (ter Haar, 1944; Kramer and ter Haar, 1946;

van de Hulst, 1949; Hoyle and Wickramasingle, 1962; Donn, 1976) have been attempted. The papers listed indicate the complexity and consequent limitations of a kinetic analysis.

#### THERMODYNAMIC EQUILIBRATION CALCULATIONS

The simplicity of thermodynamics, in which the equilibrium composition and phases in a system depend only on the final state, allows detailed results to be calculated. A determination of the chemical composition of the gas and any condensates require knowledge only of the free energies of all species which may occur. Procedures for obtaining the data and carrying through the calculations are given in the review of Grossman and Larimer (1974). They adopted a cloud of "solar" composition at a pressure of  $10^{-4}$  atmospheres. Figure 1, taken from that paper, summarizes their condensation computations. The chart shows the fraction of each element that has condensed and the mineral in which it occurs, as a function of temperature. Of the three methods for treating condensation only thermodynamics permits such detailed and comprehensive results to be obtained. Indeed, for most elements, it is the only procedure that yields any results at the present time.

Unfortunately, there is a price that must be paid for this information; the system must be in thermodynamic equilibrium. To the degree that the system is out of equilibrium, calculated results will deviate from the actual values. Several factors exist which may lead to disequilibrium. The most important among these appear to be the low pressures in clouds, and formation of metastable species which may persist indefinitely. The effect of each of these factors will be accentuated by the non-existence in the gas of the nominal molecule of the condensed mineral.

Condensation is the tendency of the system to attain equilibrium. As with many chemical reactions an activation barrier exists which may cause a non-equilibrium state to be "frozen-in". With vapor phase transitions, the requirement for supersaturation makes condensation an inherently non-equilibrium process which can have significant effects (Blander and Katz, 1968; Blander and Abdul Gawad, 1969, Blander, 1971). The experimentally determined supersaturation for iron (Frurip and Bauer, 1977) is 20 at 2000 K and increases to 500 at 1650 K. The theoretically and experimentally well established need for supersaturation thus introduces a major deviation from equilibrium. For condensation of iron at 1600 K, the iron vapor has to be undercooled by 300°. The kinetic effects of low pressure and possible metastable species will be considered in Section IV where an analysis of the kinetics of condensation is treated. It is to be emphasized that classical thermodynamics predicts the equilibrium composition and phase distribution and not the results of a phase transition which, for the reasons indicated, may yield non-equilibrium, metastable products.

### III. NUCLEATION THEORY

The essence of nucleation theory is the transition from the gaseous to the condensed phase, liquid or solid. In its usual formulation (Frenkel, 1955; Hirth and Pound, 1962; Abraham, 1974) it is a combination of thermodynamics and kinetics. The process can be represented by the set of equations:

$$A_1 + A_j \rightleftharpoons A_{j+1} \quad J = 1, 2, \dots, j^* \quad (1)$$

where  $A_1$  is the monomer,  $A_j$ , a cluster of  $j$  molecules or atoms and  $A_{j^*}$  the critical cluster or condensation nucleus. The distribution  $n(A_j)$ , for  $j$  not too near  $j^*$ , is given by thermodynamics. Monomeric addition of an existing gaseous molecule is generally assumed, although, in some cases, e.g., carbon, dimers and trimers may be involved. Only one type of condensible molecule occurs and a cluster changes size only by collisional capture or evaporative loss. Several treatments of binary systems have recently been made (Kiang et al., 1973; Shugard et al., 1974; Wilemski, 1975). Kiang and Cadle (1975) treated ternary systems. In these analyses multicomponent particles form without reaction. Condensation with reactions to form grains for which the molecular composition is different from that of the vapor has been considered explicitly by Courtney (1964) and in a more general but formal sense by Hirschfelder (1974). Characteristics of clusters which occur in these two investigations are not known, preventing application to actual systems.

Salpeter (1974) proposed that polymers incident on a growing cluster may fragment through release of the binding energy of the particle. Part is captured and the escaping fragment carries away the excess energy, stabilizing the now larger cluster. This may be an important suggestion as we shall see later.

For the formation of the most stable major minerals e.g.,  $Al_2O_3$ ,  $CaTiO_3$  (Grossman and Larimer, 1974), nucleation is expected to be seriously inhibited. The building blocks are small molecules e.g.,  $Al_2O$ ,  $TiO$ ,  $CaO$ , not the larger, nominal molecule of the crystal.



The nominal molecules yielding very stable macroscopic crystals cannot exist until clusters of many molecules occur to form the bulk material possibly in a very poor crystalline array.

At the low pressures in astronomical systems two additional factors enter. At a total pressure of  $10^{-4}$  atmos, the partial pressure of condensible species will be below  $10^{-9}$  atmos. The interval between successive collisions of condensible species with a small cluster will be greater than one second whereas the interval for all molecular collisions will be about  $10^{-5}$  s. Therefore, the rate of cluster growth is slow on molecular time scales and the stability of small clusters becomes an important factor. The binding energy per atom for metal clusters has been studied theoretically by several investigators and one experimental measurement for iron exists (Freund and Bauer, 1977). The results are summarized in Figure 2 where it is seen that the binding energies of very small clusters is about 20% of that for the bulk metal. No data exists for oxides. The stability and hence concentration of these clusters will be much less than would be found using the bulk binding energies.

A second important effect of low pressure is a marked deviation from thermal equilibrium. The consequences for chemical reactions in the interstellar medium was pointed out by Donn (1973). An analysis of non-equilibrium molecular vibrational energy in late type giant and supergiant stars was discussed by Thompson (1973). His criterion for a non-equilibrium distribution, radiative relaxation rate equal to collisional rate, is qualitative and very conservative. However, we will use the same simple measure for deviation from equilibrium here.

At  $p = 10^{-4}$  atmos, the molecular collision frequency is  $10^5 \text{ s}^{-1}$ . The efficiency of translational to vibrational energy exchange is very poor (Cottrell and McCoubrey, 1961; Kondratev, 1964a), for simple molecules at  $\sim 1000^\circ\text{K}$  an efficiency of  $10^{-3}$  may be adopted, i.e.,  $10^3$  collisions are required for energy transfer to occur. This yields a transition probability for collisional excitation of about  $100 \text{ s}^{-1}$ . Representative radiative vibrational transition probabilities are in the range 100-1000/sec. mol. (Penner, 1959). Therefore, significant deviations from a Boltzmann equilibrium distribution in the sense of a deficiency in excited states will appear at  $10^{-4}$  atmospheres.

In summary, nucleation theory makes the following assumptions:

(1) pressures are high enough for thermodynamic equilibrium to exist and for the time scale of nucleation to be short,  $\sim 10^{-5} \text{ s}$ , (2) there are unreactive, condensible species, (3) the condensed molecule exists in the vapor, (4) no reactive species exist. As we have shown, none of these assumptions apply to astronomical systems.

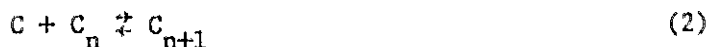
Even with the simplified approximations, nucleation theory has only been extensively used for graphite (Kamijo, 1966; Donn et al., 1968; Walker, 1975, Tabak et al., 1975). Mineral condensation sequences based on undercooling required by the large supersaturations have been derived by Blander and his associates in the references given earlier.

#### IV. KINETIC MECHANISMS

The well known distinction between thermodynamics and kinetic theory shows up in the astronomical condensation problem. Whereas thermodynamics can make detailed calculations without needing detailed processes and data as long as equilibrium exists, kinetics requires no special conditions to carry through its calculations. The price to do this

is a complete knowledge of all important processes, energies, cross-sections and other relevant data. The consequence is that calculations for specific condensates have been attempted only for ice mixtures (van de Hulst, 1949) and graphite (Hoyle and Wickramasinghe, 1962). An analysis which examined for the first time the formation kinetics of diatomic molecules as the initial step in grain formation was given by ter Haar (1942) and Kramers and ter Haar (1946). An amorphous particle consisting of essentially all elements was proposed. Cernuschi (1947) reconsidered some of the basic steps in grain formation after the initial clusters formed. In 1962, Hoyle and Wickramasinghe discussed graphite formation in the atmosphere of cool carbon stars. They made use of experimental values of the heat of reaction of small carbon polymers and theoretical value for the equilibrium vapor pressure of carbon.

What we wish to emphasize here, with regard to these investigations, is the difficulty, initially, of correctly writing down the series of processes that occur. Following this, the serious problem of obtaining energies, cross-sections and rates required for the calculation must be resolved. In the papers mentioned, mechanisms were highly simplified. In spite of the large concentration of reactive molecules, only those steps were included which added to the cluster, e.g., for graphite formation, only reactions of the form:



were included. A much more frequent step will be



where the products, CH, CH<sub>2</sub> etc. (Balooch and Olander, 1975) are produced at a rate k. Another expected reaction series is:



where H<sub>m</sub> may be an unstable fragment which yields H + H<sub>2</sub>. This is the type of process by which pyrolytic graphite is produced. The possible significant effects of hydrocarbon reactions on carbon condensation was mentioned by Donn et al., (1968). They neglected these mechanisms because of the lack of knowledge of how to treat them. Similar considerations apply to the analysis of ter Haar (1942) and the other kinetic studies.

Several other factors having important consequences on the kinetics of grain formation are briefly noted here. At low pressures three-body collisions which are needed to stabilize the association of small molecules are very infrequent. The ratio of two-to-three body collisions is approximately  $10^{-22} n$  (Kondratiev, 1964b) where n is the total particle density per cm<sup>3</sup>. In the pressure interval  $10^{-4}$  to  $10^{-8}$  atmospheres, this ratio drops from  $10^{-7}$  to  $10^{-11}$ . Salpeter's suggestion of fragmentation of the incident molecule can be effective, the escaping fragment carries away the excess binding energy and allows part of the incident molecule to add to a cluster with less than about eight atoms.

A second factor that may enhance condensation is the low vibrational temperature at low pressures which depletes the higher vibrational states of the molecules. The primary energy mode affecting condensation appears to be the internal energy of the molecule, cluster or surface rather than the molecular kinetic energy (McCarroll and Ehrlich, 1964; Shade, 1964). Thus, at high kinetic temperatures, the non-Boltzmann

distribution means that the system is effectively much colder. This may be the dominant factor allowing grains to form in "hot" clouds. A similar effect applicable to small aggregates which behave as bulk solids was pointed out by Arrhenius and De (1973).

Finally, a major contributor to condensation may be ionization which is well known to be very effective in causing nucleation in many systems (Hirth and Pound, 1963). A more general possible consequence of ionization on condensation in clouds has been considered by Arrhenius and Alfven (1970; see also Lehnert (1970)). These papers consider grain formation where plasma effects dominate clouds processes whereas this review is concerned with regions when ionization has limited, specific consequences.

#### V. REMARKS ON THE THEORETICAL MECHANISMS

The analysis of the three mechanism of investigating condensation is now summarized. Thermodynamics yields a precise, detailed evolution of condensation in clouds if the system were always at equilibrium. However, because of the fact that condensation is a transition process, not an equilibrium state, various factor may interfere with the equilibrium state being attained. Of particular significance are the low pressures characteristic of astronomical systems, and the large supersaturation required to drive the condensation process. Nucleation theory also suffers from the requirement for thermodynamic equilibrium. In addition, it has been developed only for very simple laboratory systems. The basic assumptions are not valid for astronomical cases. A generalization of nucleation theory, more valid for present circumstances may be made (Donn, 1976). This procedure will require much

more detailed knowledge than can readily be obtained and leads us to the third scheme - a kinetic treatment. Kinetics is valid under any circumstances if all the steps and quantities are known. For a low pressure, cosmic abundance system, we are very far from being able to carry out a detailed calculation. A simple, extreme calculation in which all refractory atoms and molecules add to the cluster yields a boundary condition grain. This was done previously (Donn, 1976) with the resultant composition  $\text{Fe}_7\text{Mg}_8\text{CaAlNi}(\text{SiO})_5$ . This procedure and result is somewhat analogous to that of ter Haar (19742) who was dealing with low temperature, diffu clouds where the total concentration was about unity. The composition for the grain just given may be compared with the thermodynamic results of Figure 1. Because of the considerable deviations from thermodynamic equilibrium, those calculations will not be accurate descriptions of the grains.

The general character of the thermodynamic results are expected to provide guidelines for the grain composition and condensation temperatures. Inhibition of nucleation will cause calculated temperatures to be upper limits. Elements, e.g., aluminum, titanium and calcium, are not expected to have distinct condensation temperatures or condense in unique crystalline minerals. The tendency may be for groups of elements to condense together over a range of temperatures in an amorphous aggregate. Solid state diffusion at high temperatures could cause annealing, leading to a multicrystalline grain (Stephens, 1978). The actual grain composition can be expected to fall between the two extremes. As pointed out in the earlier paper (Donn, 1976) a unique composition is unlikely. It is to be emphasized that the nature of the system and the factors that favor condensation indicate non-equilibrium solids will form.

## VI. CONDENSATION EXPERIMENTS

It should be clear that condensation experiments appropriate to the conditions in astronomical systems are needed. Experiments on condensation of smokes have been carried out by Lefevre (1967, 1970) who produced grains of iron, iron oxide, silicon carbide and silica by an arc discharge in argon. Kamijo et al., (1975) used heaters, arcs, and plasma jets to vaporize a variety of similar materials, again in argon. Steyer et al. (1974) used an arc to vaporize and recondense silica. In these experiments, condensation takes place under uncontrolled conditions with unknown temperatures and pressures. Except for iron oxide and silicon carbide, there is only a single condensible species. The analysis of condensation conditions in clouds in the previous sections has shown that these experiments are valuable for producing small grains of various materials and studying their properties but they do not contribute much to our understanding of astronomical condensation. Meyer (1971) has experimented with condensation on surfaces initiated by sputtering of selected targets by argon atoms. His procedure and results are difficult to relate to smoke formation in clouds. Of significance to the present analysis is Meyer's conclusion that some films formed by sputtering magnesium silicate targets could not be identified with any known magnesium silicate compound. Perhaps the experiment most pertinent to grain formation in astronomical clouds was the attempt by Doi and Hudson (1973) to nucleate graphite. They simulated a cool stellar atmosphere with an expanding beam of a hydrogen-methane mixture equilibrated at temperatures on the order of 2000 K but obtained a negative result with only an upper limit of 10 percent of carbon condensing.

Further work along this line could yield data on the kinetics and products of carbon condensation in hydrogen rich atmospheres.

Currently, there are two series of experiments on nucleating smokes from vaporized refractory materials which extend the work already referred to in the direction of being more realistic for cosmic clouds. The work of Arnold and his colleagues were reported at the symposium of Stephens and is described elsewhere in these proceedings. To date their experiments have been done at pressures above an atmosphere. At Goddard, Kenrick Day and I vaporized magnesium and silicon monoxide in separate, adjacent crucibles (Day and Donn, 1978). These were independently heated, allowing different Mg/Si ratios to be obtained in the vapor. An ambient argon atmosphere at about 2 Torr pressure could be heated to control the condensation temperature. Temperatures from 0 to 500°C were employed. The strong dependence of condensate yield on temperature, described below, indicates that the ambient temperature was the controlling parameter. Calculated partial pressure of the reactant Mg and SiO vapors were between 0.1 and 1 Torr. The smoke yield decreased as the ambient temperature was raised. At 500°C no smoke was collected unless the crucible temperatures were increased substantially, raising the reactant partial pressures. At 500°C the supersaturation for experiments in which no grains were collected is estimated to be greater than  $10^5$ . At all temperatures the grain size was a few hundred angstroms. The particles were amorphous, with variable Mg/Si ratios. Their infrared spectra showed structureless 10 and 20  $\mu\text{m}$  features very similar to those found in astronomical sources. When the grains were annealed in vacuum at 1000°C for 1 hour, a well developed forsterite ( $\text{Mg}_2\text{SiO}_4$ ) spectrum



developed. Figure 3 shows the infrared spectrum of condensed and annealed grains. The dependence of smoke yield on temperature, the high supersaturation and the amorphous, non-stoichiometric character of the grains, all support the kinetic mechanism for grain formation. These experiments are being extended to include other metals and more complex mixtures as well as ionization and other phenomena. The experiments will also be made more quantitative.

The concept of organic grains proposed by Hoyle and Wickramasinghe (1977) has not been considered here. The kinetic treatment is expected to apply to any composition.

The ambitious, ultimate goal of our investigation of grain formation at Goddard is to develop a method to permit approximate but realistic calculations of condensation in astronomical systems.

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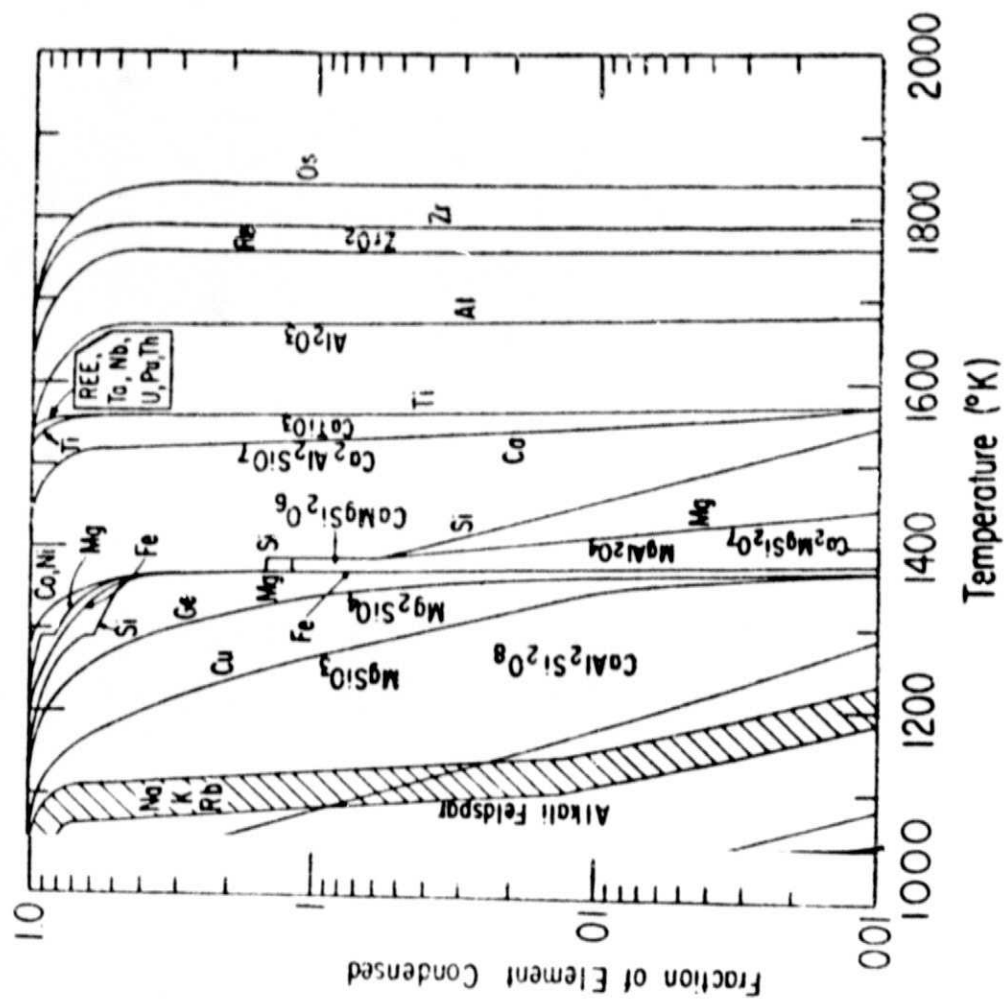
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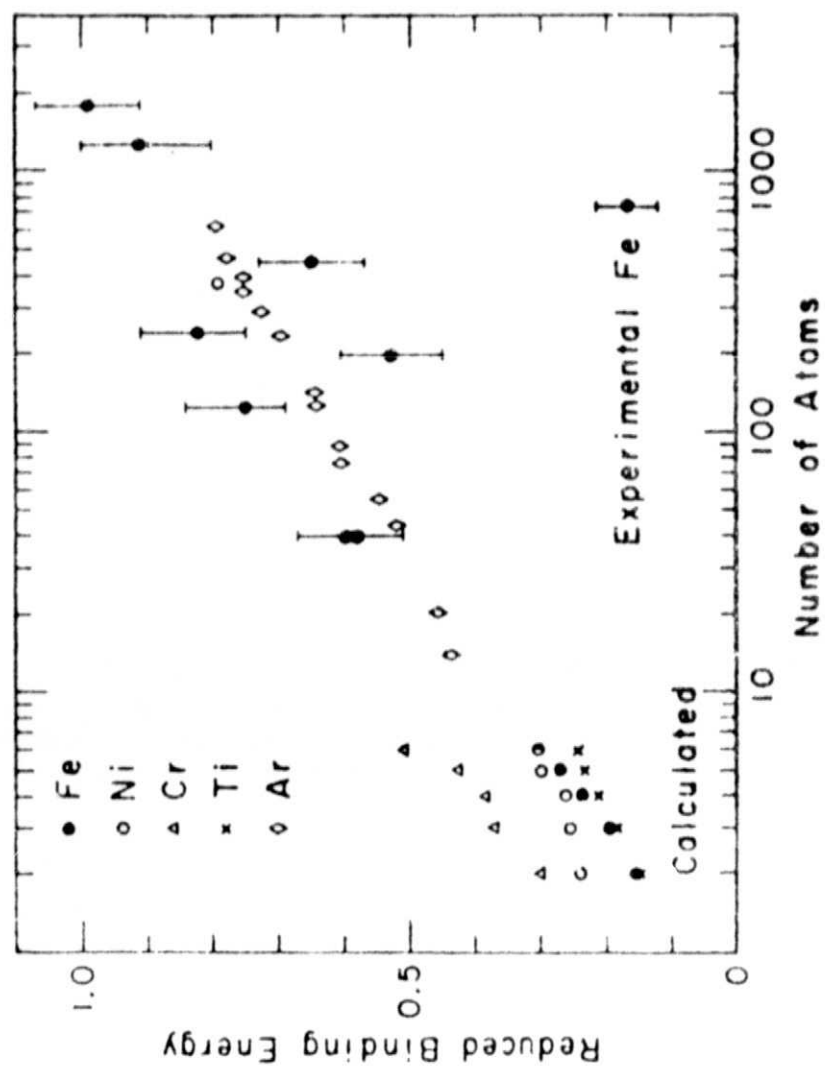
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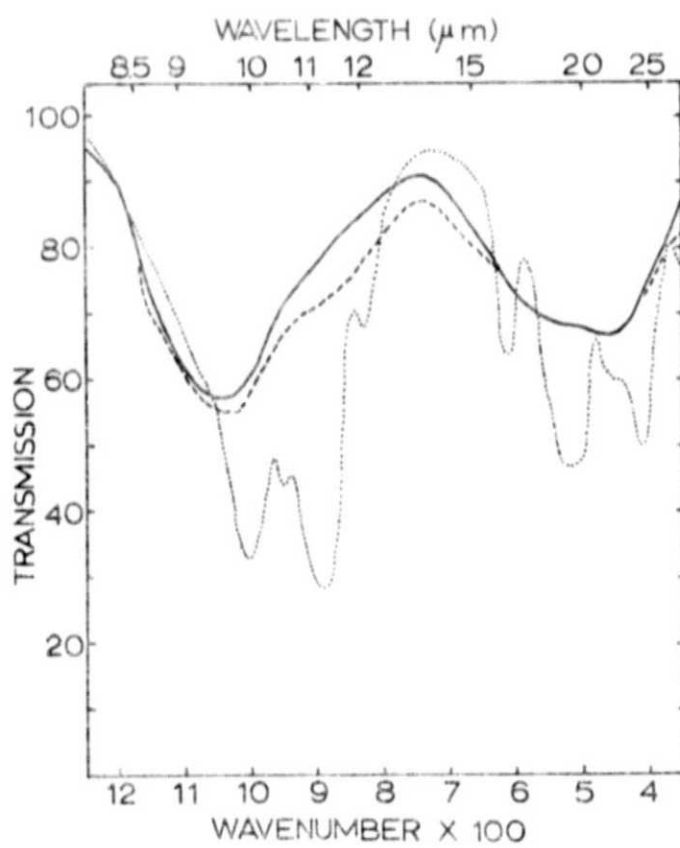
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## FIGURE CAPTIONS

- Figure 1. Summary of thermodynamic equilibrium calculations from a nebula of solar composition,  $P = 10^{-4}$  atmos. (from Grossman and Larimer, 1974).
- Figure 2. Binding energy of clusters normalized to bulk solids (from Anderson, J. Chem. Phys. 64, 4046, 1976).
- Figure 3. Infrared spectrum of smoke condensed from SiO + Mg mixture at 300 K. ---- initial condensate; \_\_\_\_\_ heated to 800 K in vacuum; .... heated to 1300 K in vacuum.









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